1396

M-CN-2047 Cy 6a

[redacted]

Metallurgical Project

A. H. Compton, Project Director

* * *

Clinton Laboratories

M. D. Whitaker, Director

R. L. Doan, Associate Director for Research

* * *

ENGINEERING DEVELOPMENT SECTION

M. C. Leverett, Section Chief

* * *

DISTRIBUTION OF FISSION ACTIVITY IN METAL WASTE SOLUTIONS

* * *

J. T. Weills and J. A. Lane

September 13, 1944

* * *

This work has been done as a necessary part of Problem Assignment 227-X16E - "Recovery of Uranium from Active Waste Solutions"

Received Clinton: 9/18/44 Series A Issued: 9/19/44
Received Chicago: Series B Issued:

Table of Contents

ABSTRACT

The 360,000 gallons of waste solution under consideration contain 77.3 tons of metal from the first 290 days operation of the Clinton 200 Area. It is stored in waste storage tanks W-10, W-7, and a small amount of W-9. That in W-9 will be mixed with an equal volume of fresh waste and the mixture used to fill W-7. The distribution of fission product activity in these tanks, important for a consideration of metal recovery, has been estimated from the fission yields and decay curves of the individual elements combined with a day by day survey of the metal discharged from the pile.

Allowing for the fact that incomplete separation from Zr, Cb, Ce and Pr is obtained in the first product precipitation, the results indicate that the activity in the waste tanks as of September 1, 1944 is the same as that from "average" metal irradiated at 15 KW per ton for 100 days, and allowed to decay 130 days. The distribution of activity and rate of decay are moreover similar to such metal. The calculated activities of the individual fission products expressed as curies and beta and gamma watts for additional decay times are summarized in Tables 2 and 3.

INTRODUCTION

A knowledge of the distribution of fission products in the metal waste solution is important for a consideration of methods of recovering the metal. After two years cooling, for example, 96% of the gamma activity is due to 36 year Cs. The best method of separating the uranium from the fission product activity at such a time would be to precipitate the uranium leaving the Cs in solution. For shorter cooling times, however, other methods of decontamination such as solvent extraction might be more efficient. As an alternate to analyzing the waste for the various fission products, the distribution of activity may be estimated from the fission yields and decay curves of the individual elements. Such curves are available from MUC-NS-212 (Project Handbook Chapter III, Section D6).

Values of pile operating power, irradiation and decay times necessary to apply the curves are found from a day by day survey of the metal discharged from the pile. These calculations have been carried out as follows:

Tracing the Source of Metal

According to the usual procedure, the metal is discharged from the pile into marked buckets for storage in the canal. Before it is sent to the 200 Area, it is redistributed in other buckets to give a more uniform concentration of product in each batch. This mades it difficult to trace the source of metal from 200 Area records. By assuming, however, that all the metal discharged from the pile up to a certain date was processed and stored in the waste storage tanks, the history of the metal may be traced from Building 105 "Detail Charging and Discharging Records." These records give the charging and discharging dates and the amount of product associated with each batch calculated on the basis of 24,000 KW hours per gram 49. This allows determination of irradiation and decay times as well as pile operating level for any given metal. By the time they were full, tanks W-9 and W-10 contained all metal discharged from the pile up to July 5, 1944 (plant runs 1 to 202) plus four/sevenths of the metal discharged from July 5 to July 17, 1944 (runs 202 to 208). The remainder of the waste from these latter runs is still held in small storage tank W-4.

After W-9 and W-10 were filled, the waste in W-9 was partially transferred to W-7. The 4' of liquid remaining in W-9 will be mixed with an equal amount of fresh waste from W-4 and the mixture used to fill W-7. The activity of the W-7 waste must, therefore, be based on a forecast of the activity of the fresh waste which will be added.

To be sure that all of the metal pushed from the pile from the beginning of operations to July 17 was processed in the 200 Area, a product balance as of August 12 was used.

This showed that the amount delivered by August 12 (from daily reports) plus amount delivered during August 12 to August 28 plus the canal inventory was within about 1% of the amount pushed, according to "Detail Charging and Discharging Records."

Calculations

Having determined the irradiation and decay times for each batch of metal, the power level may be found from the amount of 49 in the metal and the time of irradiation. Thus,

$$P = \frac{W}{T} \times 10^6$$

where, P = power level, watts.
W = weight of product, gms.
T = time in pile, days.

Activities of the individual fission elements in curies or beta and gamma watts as a function of P, T and t (the decay time) are next found from data in MUC-NS-212 or Project Handbook, Chapter III, Section D6. The activities of the Zr, Cb, Ce and Pr must be corrected for incomplete decontamination in the first product precipitation. Only 75% of the Zr and 90% of the Cb, Ce and Pr activities go into the metal waste.

Comparison of Calculated Activity with Measured Activity

Measured activities as of September 1, 1944:

α- 95 c/m/cc
β- 3.67×10^6 c/m/cc
γ- 1.5×10^4 c/m/cc

Assuming 8% geometry, 100% efficiency for beta counts, 1% efficiency for gamma counts gives for beta

$$\frac{3.67 \times 10^6 \times 180000 \times 3785}{(.08)\ (3.7 \times 10^{10})\ 60} = 14000 \text{ beta curies}$$

for gamma

$$\frac{1.5 \times 10^4 \times 180000 \times 3785}{.08 \times .01 \times 3.7 \times 10^{10} \times 60} = 5700 \text{ gamma curies}$$

This compares to calculated values of 28000 and 14000 curies for beta and gamma respectively.

Results

The total amounts of each fission element in the waste tanks on September 1, 1944 are given in Tables 2 and 3 as well as activities at future dates. A comparison of W-10 activities with values calculated on the basis of "average" metal irradiated at 15 KW per ton for 100 days and allowed to decay 130 days is given in Table 4. The calculated activities are higher for the Zr, Cb, Ce and Pr by amounts expected from the specific decontamination data in the report by W. Q. Smith, 5/1/44, "Handling of Metal Wastes."

Measured total activities are about 50% of calculated total activities.

Table 2

Activity of Fission Species in W-10 Storage Tank*

Species	Curies as of						β Watts as of						γ Watts as of					
	9/1/44	3/1/45	9/1/45	3/1/46	9/1/47	9/1/49	9/1/44	3/1/45	9/1/45	9/1/46	9/1/47	9/1/49	9/1/44	3/1/45	9/1/45	9/1/46	9/1/47	9/1/49
Sr^{89} 55d	3000	300	30	0	0	0	12.1	1.2	0.1	0	0	0	0	0	0	0	0	0
Sr^{90} 30y	200	200	200	190	190	180	0.3	0.3	0.3	0.29	0.28	0.27	0	0	0	0	0	0
Y^{90} 60h	200	200	200	190	190	180	1.0	1.0	1.0	0.96	0.94	0.90	0	0	0	0	0	0
Y(92) 57d	3400	350	40	0	0	0	11.0	1.1	0.1	0	0	0	0	0	0	0	0	0
Zr^{93} 65d	3600	550	80	2	0	0	3.7	0.6	0.1	0	0	0	16.2	2.5	0.4	0.01	0	0
Cb^{93} 35d	7300	730	100	2	0	0	6.3	0.6	0.1	0	0	0	31.8	3.2	0.4	0.01	0	0
Ru^{103} 40d	1200	60	3	0	0	0	0.8	0	0	0	0	0	3.5	0.2	0	0	0	0
Ru 330d	300	200	140	70	30	7												
Rh 30s	300	200	140	70	30	7	3.2	2.2	1.6	0.7	0.33	0.07	0	0	0	0	0	0
Te^{129} 32d																		
Te^{129} 72m	140	3	0	0	0	0	0.3	0	0	0	0	0	0.1	0	0	0	0	0
Cs^{135} 36y	150	150	150	140	140	135	0.3	0.3	0.3	0.29	0.28	0.27	0.5	0.5	0.5	0.43	0.43	0.41
Ba^{140} 12.5d	200	0	0	0	0	0	0.6	0	0	0	0	0	0	0	0	0	0	0
La^{140} 40h	200	0	0	0	0	0	0.9	0	0	0	0	0	2.5	0	0	0	0	0
Ce^{141} 28d	1400	35	0	0	0	0	2.2	0	0	0	0	0	1.9	0	0	0	0	0
Pr^{143} 13.5d	240	0	0	0	0	0	0.5	0	0	0	0	0	00	0	0	0	0	0
Ce 340d	3100	2100	1400	700	340	80												
Pr 17m	3100	2100	1400	700	340	80	22.5	15.5	10.6	5.1	2.5	0.57	0	0	0	0	0	0
TOTALS	28000	7200	3900	2100	1300	670	65.7	22.8	14.2	7.3	4.3	2.1	56.5	6.4	1.3	0.45	0.43	0.41

*Contains 180,000 gallons of waste (37.8 tons of metal).

Table 3

Estimated Total Activities in W-7 Storage Tank*

Species		Curies as of 3/1/45	Curies as of 9/1/45	β Watts as of 3/1/45	β Watts as of 9/1/45	γ Watts as of 3/1/45	γ Watts as of 9/1/45
Sr^{89}	55d	1000	100	4.0	0.4	0	0
Sr^{90}	30y	300	300	0.4	0.4	0	0
Y^{90}	60h	300	300	1.6	1.6	0	0
$Y^{(92)}$	57d	1000	100	3.6	0.4	0	0
Zr^{93}	65d	2000	300	1.9	0.3	8.6	1.2
Cb^{93}	35d	2500	300	2.2	0.3	10.4	1.5
Ru^{103}	40d	250	10	0.2	0	0.9	0
Ru	330d	800	500	4.0	2.8	0	0
Rh	30s	800	500			0	0
Te^{129}	32d	20	0	0.1	0	0	0
Te^{129}	72m						
Cs^{135}	36y	250	250	0.4	0.4	0.7	0.7
Ba^{140}	12.5d	1	0	0	0	0	0
La^{140}	40h	1	0	0	0	0	0
Ce^{141}	28d	200	2	0.3	0	0.2	0
Pr^{143}	13.5d	1	0	0	0	0	0
Ce	340d	4000	3000	30.8	21.3	0	0
Pr	17m	4000	3000			0	0
Totals		30000	10000	50	30	20	4

*Will contain when full 180,000 gallons of waste (38.7 tons metal).

Table 4

	Activity of fission species in W-10 as of September 1, 1944.			Activity of fission species calculated on basis of metal reacted at 15 MW/ton for 100 days, cooled 130 days.		
Element	Curies	Watts β	Watts γ	Curies	Watts β	Watts γ
55d Sr	3000	12.1	0	3000	12.1	0
30y Sr	200	0.3	0	180	.25	0
50h Y	200	1.0	0	180	.9	0
57d Y	3400	11.0	0	3300	10.9	0
65d Zr*	3600	3.7	16.2	4700	4.7	21.4
35d Cb*	7300	6.3	31.8	8800	7.5	37.4
40d Ru	1200	0.8	3.5	1000	0.7	2.8
33d Ru	300			360		
30s Rh	300	3.2	0	360	3.5	0
32d(72m) Te	140	0.3	0.1	120	.25	.06
36y Cs	150	0.3	0.5	120	.38	.48
12.5d Ba	200	0.6	0	30	.05	0
40h La	200	0.9	2.5	30	.10	.34
28d Ce	1400	2.2	1.9	1050	1.4	1.21
13.5d Pr	240	0.5	0	30	.07	0
34Cd Ce*	3100			3600		
17m Pr*	3100	22.5	0	3600	26.8	0
TOTAL	28000	65.7	56.5	30400	69.6	63.7

*The Zr, Cb, Ce and Pr activities in the waste are lower than calculated due to partial precipitation of these elements with the first product precipitation.